

Low temperature bonding of heterogeneous materials using Al₂O₃ as an intermediate layer

Sahoo, Hitesh Kumar; Ottaviano, Luisa; Zheng, Yi; Hansen, Ole; Yvind, Kresten

Published in:

Journal of Vacuum Science and Technology. Part B. Microelectronics and Nanometer Structures

Link to article, DOI:

[10.1116/1.5005591](https://doi.org/10.1116/1.5005591)

Publication date:

2018

Document Version

Peer reviewed version

[Link back to DTU Orbit](#)

Citation (APA):

Sahoo, H. K., Ottaviano, L., Zheng, Y., Hansen, O., & Yvind, K. (2018). Low temperature bonding of heterogeneous materials using Al₂O₃ as an intermediate layer. Journal of Vacuum Science and Technology. Part B. Microelectronics and Nanometer Structures, 36(1), [011202]. DOI: 10.1116/1.5005591

DTU Library

Technical Information Center of Denmark

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

Low temperature bonding of heterogeneous materials using Al_2O_3 as an intermediate layer

Hitesh Kumar Sahoo,^{1, a)} Luisa Ottaviano,¹ Yi Zheng,¹ Ole Hansen,² and Kresten Yvind^{1, b)}

¹⁾*DTU Fotonik, Technical University of Denmark, 2800 Kongens Lyngby, Denmark*

²⁾*DTU Nanotech, Technical University of Denmark, 2800 Kongens Lyngby, Denmark*

Integration of heterogeneous materials is crucial for many nano-photonics devices. The integration is often achieved by bonding using polymer adhesives or metals. A much better and cleaner option is direct wafer bonding, but the high annealing temperatures required makes it a much less attractive option. Direct wafer bonding relies on a high density of hydroxyl groups on the surfaces, which may be difficult to achieve dependent on the materials. Thus, it is a challenge to design a universal wafer bonding process. However, using an intermediate layer between the bonding surfaces reduces the dependence on the bonding materials and thus the bonding mechanism essentially remains the same. We present a systematic study on the use of Al_2O_3 as an intermediate layer for bonding of heterogeneous materials. The ability to achieve a high hydroxyl group density and well-controlled films makes atomic layer deposited (ALD) Al_2O_3 an excellent choice for the intermediate layer. We have optimized the bonding process to achieve a high interface energy of 1.7 J/m^2 for a low temperature annealing of 300°C . We also demonstrate wafer bonding of InP to SiO_2 on Si and GaAs to sapphire using the Al_2O_3 interlayer.

PACS numbers: Valid PACS appear here

Keywords: heterogeneous materials, direct bonding, Al_2O_3 , low temperature, Silicon photonics, laser

I. INTRODUCTION

Direct wafer bonding is a key enabling technology for many current and emerging nanophotonic devices. Most of the work on direct wafer bonding has however been focused on the Si platform for fabrication of silicon-on-insulator (SOI) and micro-electromechanical systems (MEMS). The work on direct wafer bonding of Si to Si/ SiO_2 using high temperature annealing from Maszara et al.¹ is one of the earliest efforts on understanding bonding. Tong et al.² furthered the understanding by investigating the contribution of hydroxyl groups and achieved a high interface energy by increased annealing time. These works remain the gold standards for direct bonding. Adapting the bonding process to heterogeneous materials is, however, a challenge. Firstly, high temperature annealing of hetero bonded samples gives rise to thermal stress in the film or even defects due to mismatched coefficients of thermal expansion. For example, annealing of InP bonded to Si at a temperature above 300°C leads to a build-up of internal stresses in the bonded wafers which creates defects in the InP wafer.³ Integration of heterogeneous materials thus remains one of the roadblocks for semiconductor devices on new platforms such as III-V on Si, Ge on Sapphire, LiNbO_3 on GaAs etc. Secondly, it is important to maximize the density of bond sites, i.e., the hydroxyl group density for a strong bond. A common way to achieve this is to break the surface bonds by O_2 plasma, Ar sputtering, etc. on a wafer followed by a dip in de-ionized (DI) water to re-hydrolyze the surface. This method has a few drawbacks and cannot be adapted to all systems. The process of breaking surface bonds increases surface roughness.^{4,5} Also, dipping in water is not desired, especially when you have MEMS and there is a risk of damaging them. In addition, the maximum hydroxyl group density on a wafer

surface is an intrinsic property of the material and thus cannot be increased beyond a certain limit for the desired material. Thus, adapting the Si-Si bonding process to a new material platform does not necessarily guarantee an acceptable bond quality. These concerns have generated a strong interest in finding an alternative solution for bonding of heterogeneous materials at low temperature.

The key to achieving a high interface energy at low temperature lies in the choice of a material with high hydroxyl group density and use that material as an intermediate layer for direct bonding. This would increase reliability and reduce dependence on the substrate material for achieving a high interface energy. Al_2O_3 is an excellent choice for an intermediate layer for direct bonding. It has a high hydroxyl group density of around 18 OH/nm^2 ,^{6,7} more than 4 times higher than that of Si.^{8,9} Al_2O_3 can also be deposited with high accuracy using atomic layer deposition (ALD). The ALD process helps in achieving thickness control at the sub-nm scale and does not add any surface roughness. There has been some reports on Al_2O_3 ^{10–13} based bonding being stronger than Si/ SiO_2 direct bonding. However, there is a lack of systematic studies to understand the bonding mechanism using Al_2O_3 as the intermediate layer. In this work, we have investigated the contribution of deposition parameters for Al_2O_3 , the thickness of Al_2O_3 , and annealing conditions to the bond strength. Finally, an optimized process flow for bonding heterogeneous materials is presented; the process can be applied to most material systems.

II. METHODOLOGY

Characterization is important for the ability to quantify a good bond. It can be either a quantitative or qualitative measurement. Common quantitative wafer bond characterization methods include Maszara's blade test,¹ and micro-chevron test.¹⁴ Despite high reliability of the latter, the Maszara test is the most used method,^{15–18} because of its simplicity and ease

^{a)}Electronic mail: hikus@fotonik.dtu.dk

^{b)}Electronic mail: kryv@fotonik.dtu.dk

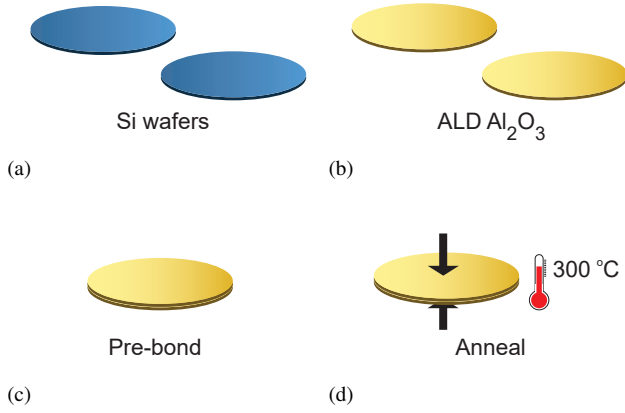


FIG. 1: (Color online) Illustrations of the basic steps for bonding wafers using Al_2O_3 as an intermediate layer. (a) Two prime grade 2 inch clean Si wafers with surface roughness around 0.2 nm were chosen in this experiment. (b) A thin layer of ALD Al_2O_3 was deposited on both wafers. (c) The wafers were then placed on top of each other for pre-bonding. An additional exposure to water vapor, before the pre-bonding, was done for one experiment run to understand the effect of hydroxyl density and temperature. (d) The wafers were finally annealed at low temperature ($\leq 300^\circ\text{C}$) and with a controlled applied pressure.

of use. Maskteika et al.¹⁹ has identified some of the common mistakes and articulated the correct methodology for an accurate Maszara test. Even so, brittleness of some of the wafers (InP, GaAs, etc.) makes it a challenge to do any of the above-mentioned tests. On the other hand, qualitative measurement refers to the ability of the bonded sample to survive polishing or substrate removal etch. It does not sacrifice the sample, unlike the quantitative methods. So, for expensive wafers like InP, GaAs, Sapphire, LiNbO_3 , etc., the most common characterization method has been qualitative. This has resulted in an incomplete knowledge of the bond quality and the mechanism behind a successful bonding. Thus, to be able to characterize and understand Al_2O_3 based direct bonding, Si wafers were chosen for the experiment. This helped us benchmarking our approach against the standard Si direct bonding with high annealing temperatures. Bonding is primarily a surface phenomenon and depends entirely on the forces between the bonding surfaces. Apart from stress from the wafer, there is almost no contribution from the underlying material. Thus, any result obtained from bonding of Si wafers using Al_2O_3 as the intermediate layer can be adapted to most material systems.

The basic bonding steps are illustrated in the Figure 1. Two prime grade single crystal (100) 2-inch clean Si wafers with a surface roughness of around 0.2 nm were used for each run. The wafers were taken out of a new sealed wafer box and ALD Al_2O_3 was deposited on both wafers. The deposition process was done in a commercial thermal ALD system using liquid precursors Trimethyl Aluminum (TMA) and de-ionized water (DIW). A pulse time of 0.1 s was used for both TMA and H_2O with a nitrogen flow of 150 sccm and 200 sccm respectively. The thickness of the deposited film was controlled by the number of cycles of ALD. The temperature for deposition

was kept between 200°C and 300°C in-order to achieve a uniform and high quality deposition of Al_2O_3 .^{20–22} The deposition rate of $\sim 1 \text{ \AA}/\text{cycle}$ was obtained in the operating temperature regime. The wafers were pre-bonded right out of the chamber. The wafers were placed on top of each other with the bondable surface facing each other and a small force was applied in the center of the wafers for initiating the bond wave. In some runs, the wafers were exposed to water vapor for 30 sec before pre-bonding, to study the effect of hydroxylation. The pre-bonded wafers were then annealed. The annealing step was done in a commercial wafer bonder. The annealing temperature was varied between 200 and 300°C . The annealing temperature is specific to the materials being bonded. For example, the maximum allowed temperature for annealing an InP wafer bonded to Si would be 300°C . An annealing time of 60 min was used for all runs except when the effect of annealing time was studied. A force of 2 kN was applied on the 2 inch bonded wafers during the annealing process. To investigate the contribution from applied pressure during annealing, some samples were also annealed in a furnace without any applied force. The bonded wafers were characterized using the Maszara blade test in a cleanroom atmosphere by measuring the de-lamination from the insertion of a $50 \mu\text{m}$ thick steel strip between the wafers. The interface energy, γ was then calculated using¹

$$\gamma = \frac{3}{32} \frac{Et^3y^2}{L^4}, \quad (1)$$

where E is Young's modulus of the material, t is the thickness of the wafers, y is the thickness of the blade and L is the crack length. Care was taken to use the correct value of E for Si²³ based on the direction of insertion of the blade relative to the wafer flat. The experimental error for the measurement method is typically $\pm 15\%$ due to variations on the wafer surfaces and crack length measurement procedure. In the results shown below, the error bars represent the standard deviation of the measurements.

III. RESULTS

A. Thickness of ALD Al_2O_3

The amount of Al_2O_3 required for use as an intermediate layer for direct bonding is very important. It would be desirable to have as little as possible and at the same time not to compromise on the bond quality. For applications such as bonding for MEMS VCSELs²⁴ where ALD Al_2O_3 would be deposited even on the mechanical structure, it is critical to have as thin a layer as possible in order not to alter the mechanical properties of the structure. On the other hand, the amount of material should not be too small for an effective bond.

To study the amount of Al_2O_3 required for a good bond, the number of ALD cycles was varied from 25 to 500 cycles ($\sim 2\text{-}50 \text{ nm}$). The minimum number of cycles was chosen as 25 cycles to ensure a uniform deposition^{25,26} on the whole wafer. The upper limit was chosen to be 500 cycles to test bond-ability with a thick layer of Al_2O_3 . Figure 2 shows the interface energy measured for wafers bonded with different thickness of the intermediate material. For all thicknesses be-

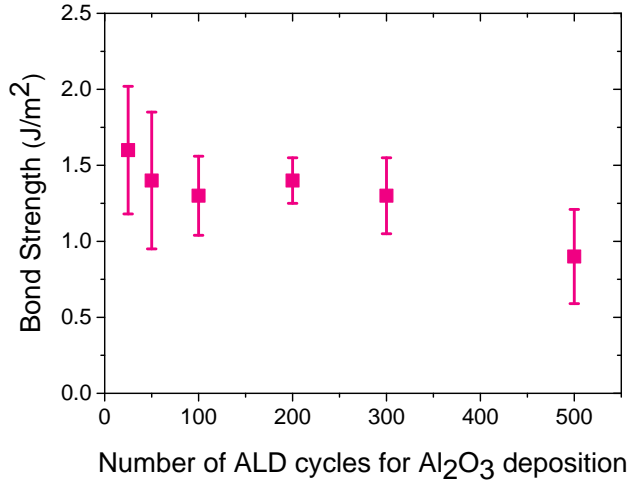


FIG. 2: (Color online) Interface energy of Si wafers bonded using different thickness of Al₂O₃ as the intermediate layer. Al₂O₃ was deposited using ALD at 200 °C. The deposition rate is ~ 1 Å/cycle. The samples were annealed at 300 °C for 60 min. The symbols represent the average interface energy and bars represent the standard deviation in the measurements.

low 300 cycles, almost no appreciable change in interface energy was observed. This agrees with our earlier discussion on bonding as a surface phenomenon. So, an intermediate layer of as thin as 2 nm Al₂O₃ should be enough for a successful bond. The samples with 50 nm of ALD Al₂O₃, however, showed slightly lower interface energy. It is remarkable that very good bonding strength is obtained in all cases, since the native oxide is known to be too thin^{27–29} to accommodate the hydrogen molecules that are usually evolved during annealing. Hydrogen is evolved due to oxidation of silicon by water arising from reaction of the surface hydroxyl groups. The hydrogen is expected to form bonding defects in the form of interface bubbles,²⁸ which reduce the bonding strength. We suggest that the native oxide (annealed in the ALD process) and the much denser Al₂O₃ film³⁰ in combination prevents diffusion of water to the silicon interface and thus hydrogen evolution is avoided. Ventosa et al.³¹ showed that annealing of the native oxide could prevent hydrogen evolution and lead to void-free bonding.

Thus, a few nm of ALD Al₂O₃ (2 nm, to ensure uniform coverage) can be used as an intermediate layer to achieve a good bond. So, an optimized 25 cycles of ALD Al₂O₃ was used for the next experimental runs.

B. Density of (-OH) groups

As mentioned earlier, high hydroxyl group density is responsible for robust bond formation as also highlighted in the literature^{2,19} on low temperature bonding mechanisms. The pre-bond step brings bondable surfaces in close proximity and a weak hydrogen bond is formed between the two surfaces. With annealing, these sites would give way to the formation of Al-O-Al thereby chemically binding the two surfaces. So, the process flow should be optimized for maximizing the hy-

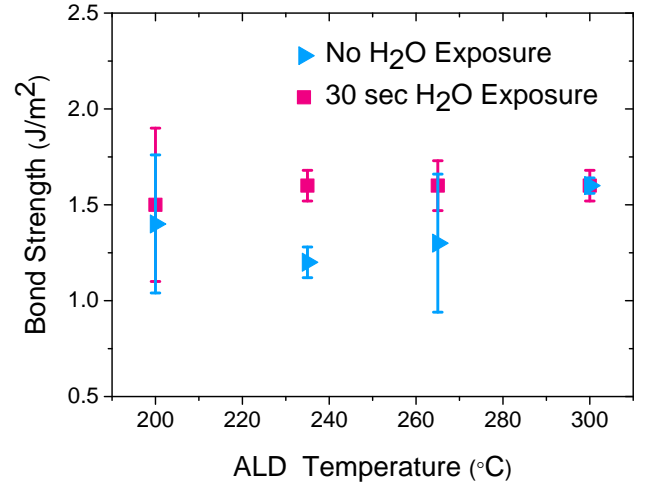
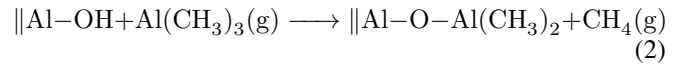


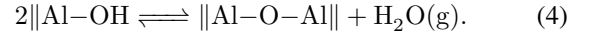
FIG. 3: (Color online) Interface energy of Si wafers bonded using Al₂O₃, deposited at different temperatures, with (squares) or without (triangles) a 30 sec H₂O exposure between unloading of wafers from the chamber and pre-bonding. All the samples were annealed at 300 °C with a force of 2 kN on 2 inch full wafers. The symbols represent the average interface energy and bars represent the standard deviation in the measurements.

droxyl group density on the wafer surface. In this case, the ALD deposition process for Al₂O₃ decides the surface chemistry. The process of ALD deposition can be described by the equations 2 and 3^{32–34}



where \parallel implies the top surface of the wafer.

The first reaction with TMA is irreversible and is not affected by the temperature within the reaction temperature regime.³⁴ The second step, hydroxylation of the surface is often mentioned without a competing reaction, which is a temperature dependent step and is a reversible reaction (Equation 4).^{20,35} At high temperatures, the hydroxy groups from the surface ($\parallel\text{Al}-\text{OH}$) may react with each other (Equation 4) and thus reduce the number of hydroxy groups on the surface



So, to test the effect of the dehydroxylation temperature dependent step, 25 cycles of ALD Al₂O₃ were deposited at different temperatures between 200 °C and 300 °C. In another set of parallel runs, the wafers were unloaded from the ALD chamber and were exposed to water vapor for 30 sec before being brought in contact with each other. This process step was included in an effort to re-hydrolyze the surface. In all the above runs, the wafers were not settled on any heat sink.

Figure 3 shows the interface energy measured for wafers bonded with 2 nm of Al₂O₃ intermediate layer deposited at different ALD temperatures. The first set of runs where the temperature of deposition was the only variable, there is no appreciable difference in the interface energy. Even though a

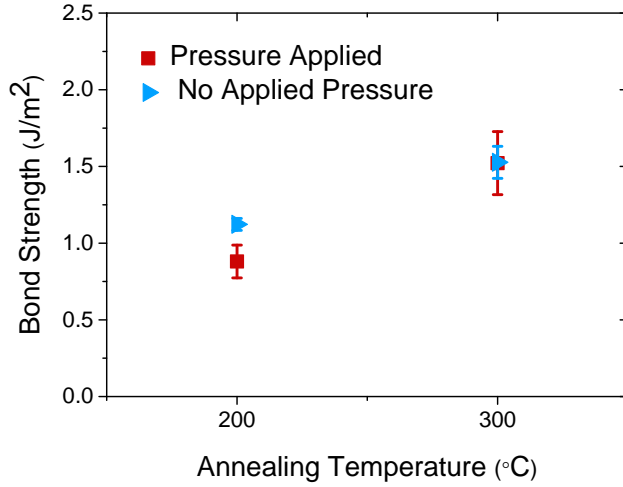


FIG. 4: (Color online) Interface energy of Si wafers bonded using Al_2O_3 as the intermediate layer and annealed at different temperatures with/without application of pressure (15 kN on 2 inch wafer) for 60 min. Increase in temperature increases the interface energy but there is no observed effect of applied pressure during annealing.

decrease in the interface energy with ALD deposition temperature is expected due to decreased hydroxyl density, the temperature regime for the experiments is most likely too narrow to show any profound effect. The second set of runs which included an added 30 sec exposure to water vapor definitely shows a small increase in interface energy relative to the first set of runs. Furthermore, the interface energy for these runs is independent of the ALD deposition temperature. Thus it can be concluded that while ALD deposition temperature has a direct influence on the hydroxyl group density on the wafers, a short exposure to an atmosphere with high relative humidity will help restore some of the hydroxyl groups. To further improve the process, ALD Al_2O_3 deposited wafers should be put on a metal plate (heat sink) to bring the wafer temperature to room temperature in an atmosphere with 50 % relative humidity (standard clean-room). This should help increase the hydroxyl group density.

C. Annealing Parameters

Annealing of bonded samples helps increase the interface energy. There are three important parameters - annealing temperature, applied pressure and annealing duration. With higher annealing temperatures, the interface water molecules become more mobile and lead to a change from hydrogen bonding towards covalent bonds. Applied pressure helps bring the wafer surfaces closer and might contribute to increasing the bond quality. Equally important is the duration of annealing, which roughly translates to the time assigned for the above-mentioned mechanism to happen.

In order to optimize the annealing parameters, the pre-bonded wafers were annealed at 200 °C and 300 °C, with/without applied pressure and the annealing duration was varied from 1 hr to 20 hr. The annealing temperature was limited to 300 °C to be compatible with heterogeneous bonding. The

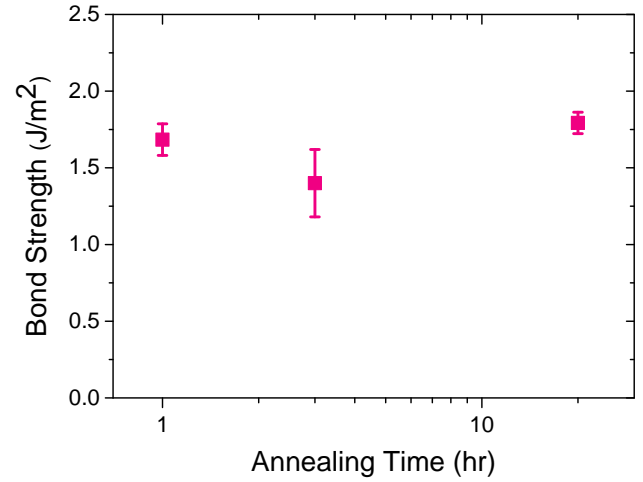


FIG. 5: (Color online) Interface energy of Si wafers bonded using Al_2O_3 deposited at 200 °C as the intermediate layer and annealed at 300 °C for different duration. There was no significant change in the interface energy when annealing time was increased from 1 hr to 20 hr.

applied pressure was limited by the maximum allowed force on the commercial wafer bonder. The annealing time was restricted to 20 hr due to limited availability of wafer bonder. Figure 4 confirms that with an increase in annealing temperature, there is an observed increase in the interface energy. Applied pressure does not have any appreciable effect on the interface energy. However, in spite of this experimental result, small particles might get trapped due to wafer handling and an applied pressure would help reduce the size of voids formed due to them. Finally, Figure 5 shows that the interface energy does not change with annealing time, at least for 20 hours duration used in this experiment.

D. Optimized process flow

The above-discussed results help in proposing an optimized bonding process using Al_2O_3 as the intermediate layer. Initially, the requirement on cleanliness and surface roughness remain almost the same as that of direct bonding, except there is no need for standard wet cleaning steps (RCA, Piranha, etc). Proper handling should be ensured to minimize contamination. Next, as deduced from Figure 2, the wafers are coated with a thin layer (~ 2 nm) of ALD Al_2O_3 , deposited at 200 °C. The wafers are bonded right after unloading from the ALD chamber with no additional activation step. The wafers may, however, be placed on a heat sink for some time to maximize the hydroxyl coverage. Lastly, the wafers are to be annealed at highest permissible temperature with an application of small force for 60 min. The bonded wafers are expected to have a high interface energy of around 1.7 J/m². The annealing time, in particular, can be further optimized to achieve a saturated interface energy as reported in Tong et al.² An extended anneal can be used after an initial anneal under pressure. Figure 6 shows the interface energy obtained in this work benchmarked against Si-Si direct bonding with annealing time of 1 hr¹ and 100 hr,² respectively. The achieved interface energy

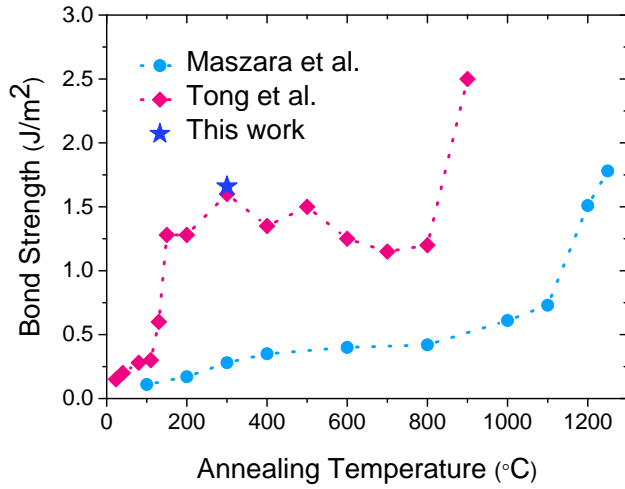


FIG. 6: (Color online) Interface energy of Si-Si wafers bonded using Al_2O_3 as intermediate layer annealed at 300 °C for 1 hr, benchmarked against direct Si-Si bonding annealed for 1 hr (Maszara et al.¹) and 100 hr (Tong et al.²).

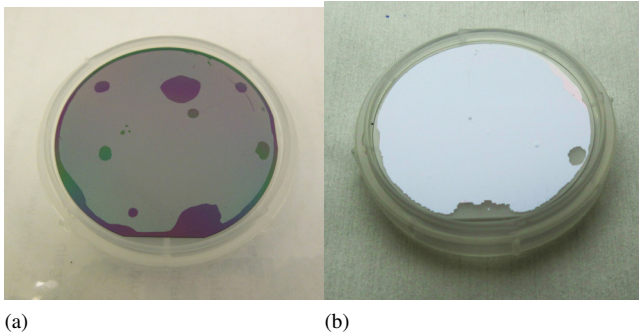


FIG. 7: (Color online) Images of 2-inch bonded wafers. (a) InP bonded to SiO_2 on Si and (b) GaAs bonded to Sapphire after a III-V substrate etch process.

was high enough for our requirements (surviving substrate removal and post-processing).

IV. BONDING HETEROGENEOUS MATERIALS

The optimized process flow, mentioned above, was successfully applied to two different hetero wafer systems. A 2 inch InP wafer was bonded to thermal oxide grown on a Si wafer and a 2 inch GaAs wafer was bonded to a sapphire wafer. After the bonding, the III-V substrates were etched away until a stop layer, thus leaving less than a micron of InP or GaAs behind (Figure 7). The ability to survive the etch is used by many experimentalists as a qualitative test for the bonding. The voids visible on the wafers are from particles on the wafers before bonding. In another run, a patterned wafer ($\sim 1\mu\text{m}$ steps of SiO_2 on a Si wafer) was bonded to InP. A cross-sectional image of the cleaved wafer is shown in Figure 8. The wafers have been processed further towards interesting nanophotonic devices. For these wafers, Maszara test could not be performed since the high interface energy made III-V

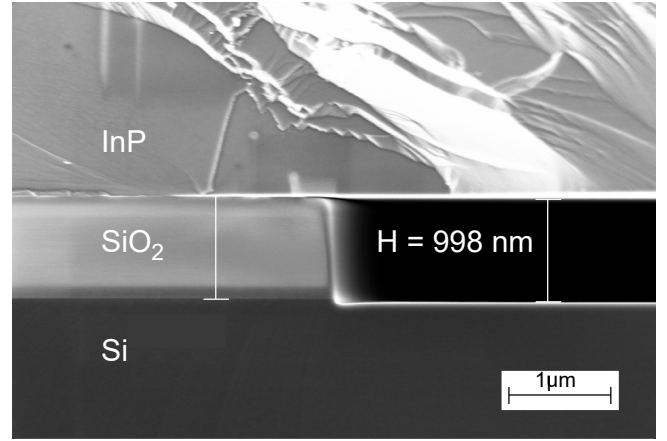


FIG. 8: Cross-section SEM image of a bonded interface. Patterned SiO_2 on Si using dry etch was bonded to InP wafer with Al_2O_3 as the intermediate layer.

wafers break instead of delamination.

V. CONCLUSIONS

We have investigated and optimized a low temperature bonding process using Al_2O_3 as an intermediate layer to bond heterogeneous material systems. Al_2O_3 is an excellent choice for the intermediate layer since it has a high hydroxyl group density. We investigated the contribution of ALD Al_2O_3 thickness, deposition temperature and annealing parameters in order to achieve a high interface energy. We also avoided wet processing or any separate activation steps. This is quite desirable for bonding wafers with open MEMS structures. An interface energy of 1.7 J/m^2 was achieved for Si-Si bonding using Al_2O_3 as the intermediate layer after annealing for 1 hr at 300 °C. The value is higher than the saturated interface energy for Si-Si direct bonding after annealing at 300 °C. Lastly, we also demonstrated adaptation of the bonding process to two hetero-material platforms.

The presented bonding process technology is expected to find application in the integration of III-V on Si, Ge on sapphire, LiNbO_3 on GaAs, etc and thus paving the way toward a new family of devices.

ACKNOWLEDGMENT

The authors would like to acknowledge the financial support from the Danish Innovation Foundation through the HERON project, the Villum Center of Excellence NATEC and the National Basic Research Foundation center SPOC (DNR123). Andrey Marchevski and Aurimas Sakanas are acknowledged for providing the image of InP bonded to Si/ SiO_2 in Figure 7.

REFERENCES

- W. P. Maszara, G. Goetz, A. Caviglia, and J. B. McKitterick, *Journal of Applied Physics* **64**, 4943 (1988).

- ²Q.-Y. Tong and U. Gösele, *Journal of the Electrochemical Society* **143**, 1773 (1996).
- ³D. Pasquariello and K. Hjort, *IEEE Journal of Selected Topics in Quantum Electronics* **8**, 118 (2002).
- ⁴U. Alam, M. M. R. Howlader, and M. J. Deen, *Journal of Micromechanics and Microengineering* **24**, 035010 (2014).
- ⁵D. Pasquariello, C. Hedlund, and K. Hjort, *Journal of The Electrochemical Society* **147**, 2699 (2000).
- ⁶A. A. Tsyganenko and P. P. Mardilovich, *Journal of the Chemical Society, Faraday Transactions* **92**, 4843 (1996).
- ⁷M. Digne, P. Sautet, P. Raybaud, P. Euzen, and H. Toulhoat, *Journal of Catalysis* **211**, 1 (2002).
- ⁸C. G. Armistead, A. J. Tyler, F. H. Hambleton, S. A. Mitchell, and J. A. Hockey, *Journal of Physical Chemistry* **73**, 3947 (1969).
- ⁹L. T. Zhuravlev, *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **173**, 1 (2000).
- ¹⁰J. Fan, L. C. Tu, and C. S. Tan, *AIP Advances* **4**, 031311 (2014).
- ¹¹Y. Li, S. Wang, B. Sun, H. Chang, W. Zhao, X. Zhang, and H. Liu, *ECS Transactions* **50**, 303 (2013).
- ¹²Y. Ikku, M. Yokoyama, R. Iida, M. Sugiyama, Y. Nakano, M. Takenaka, and S. Takagi, in *Compound Semiconductor Week (CSW/IPRM), 2011 and 23rd International Conference on Indium Phosphide and Related Materials* (IEEE, 2011) pp. 1–4.
- ¹³J. Fan, P. Anantha, C. Y. Liu, M. Bergkvist, H. Wang, and C. S. Tan, *ECS Journal of Solid State Science and Technology* **2**, N169 (2013).
- ¹⁴M. Petzold, J. Bagdahn, and D. Katzer, *Microelectronics reliability* **39**, 1103 (1999).
- ¹⁵J.-P. Raskin, *Journal of Microelectromechanical Systems* **14**, 368 (2005).
- ¹⁶G. Y. Chong and C. S. Tan, *Journal of The Electrochemical Society* **158**, H137 (2011).
- ¹⁷D. Liang and J. E. Bowers, *Journal of Vacuum Science & Technology B: Microelectronics and Nanometer Structures* **26**, 1560 (2008).
- ¹⁸M. A. Schmidt, *Proceedings of the IEEE* **86**, 1575 (1998).
- ¹⁹V. Masteika, J. Kowal, N. J. St Braithwaite, and T. Rogers, *ECS Journal of Solid State Science and Technology* **3**, 42 (2014).
- ²⁰M. Juppo, A. Rahtu, M. Ritala, and M. Leskelä, *Langmuir* **16**, 4034 (2000).
- ²¹A. Ott, J. Klaus, J. Johnson, and S. George, *Thin Solid Films* **292**, 135 (1997).
- ²²R. Matero, A. Rahtu, M. Ritala, M. Leskelä, and T. Sajavaara, *Thin Solid Films* **368**, 1 (2000).
- ²³M. A. Hopcroft, W. D. Nix, and T. W. Kenny, *Journal of microelectromechanical systems* **19**, 229 (2010).
- ²⁴H. K. Sahoo, T. Ansbæk, L. Ottaviano, E. S. Semenova, O. Hansen, and K. Yvind, in *Society of Photo-Optical Instrumentation Engineers (SPIE) Conference Series*, Vol. 9760 (2016).
- ²⁵S. Swaminathan, Y. Sun, P. Pianetta, and P. C. McIntyre, *Journal of Applied Physics* **110** (2011), 10.1063/1.3647761.
- ²⁶V. Naumann, M. Otto, R. Wehrspohn, M. Werner, and C. Hagendorf, *Energy Procedia* **27**, 312 (2012).
- ²⁷T. Plach, K. Hingerl, S. Tollabimazraehno, G. Hesser, V. Dragoi, and M. Wimplinger, *Journal of Applied Physics* **113**, 094905 (2013).
- ²⁸S. Vincent, I. Radu, D. Landru, F. Letertre, and F. Rieutord, *Applied Physics Letters* **94**, 101914 (2009).
- ²⁹C. Ventosa, F. Rieutord, L. Libralesso, C. Morales, F. Fournel, and H. Moriceau, *Journal of Applied Physics* **104**, 123524 (2008).
- ³⁰E. Langereis, M. Creatore, S. B. S. Heil, M. C. M. Van De Sanden, and W. M. M. Kessels, *Applied Physics Letters* **89**, 2 (2006).
- ³¹C. Ventosa, F. Rieutord, L. Libralesso, F. Fournel, C. Morales, and H. Moriceau, *Journal of The Electrochemical Society* **156**, H818 (2009).
- ³²C. Soto and W. Tysoe, *Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films* **9**, 2686 (1991).
- ³³A. C. Dillon, A. W. Ott, J. D. Way, and S. M. George, *Surface Science* **322**, 230 (1995).
- ³⁴R. L. Puurunen, *Journal of applied physics* **97**, 9 (2005).
- ³⁵A. Rahtu, T. Alaranta, and M. Ritala, *Langmuir* **17**, 6506 (2001).

FIGURE CAPTIONS

[FIG 1] (Color online) Illustrations of the basic steps for bonding wafers using Al_2O_3 as an intermediate layer. (a) Two prime grade 2 inch clean Si wafers with surface roughness around 0.2 nm were chosen in this experiment. (b) A thin layer of ALD Al_2O_3 was deposited on both wafers. (c) The wafers were then placed on top of each other for pre-bonding. An additional exposure to water vapor, before the pre-bonding, was done for one experiment run to understand the effect of hydroxyl density and temperature. (d) The wafers were finally annealed at low temperature ($\leq 300^\circ\text{C}$) and with a controlled applied pressure.

[FIG 2] (Color online) Interface energy of Si wafers bonded using different thickness of Al_2O_3 as the intermediate layer. Al_2O_3 was deposited using ALD at 200°C . The deposition rate is $\sim 1 \text{ \AA/cycle}$. The samples were annealed at 300°C for 60 min. The symbols represent the average interface energy and bars represent the standard deviation in the measurements.

[FIG 3] (Color online) Interface energy of Si wafers bonded using Al_2O_3 , deposited at different temperatures, with (squares) or without (triangles) a 30 sec H_2O exposure between unloading of wafers from the chamber and pre-bonding. All the samples were annealed at 300°C with a force of 2 kN on 2 inch full wafers. The symbols represent the average interface energy and bars represent the standard

deviation in the measurements.

[FIG 4] (Color online) Interface energy of Si wafers bonded using Al_2O_3 as the intermediate layer and annealed at different temperatures with/without application of pressure (15 kN on 2 inch wafer) for 60 min. Increase in temperature increases the interface energy but there is no observed effect of applied pressure during annealing.

[FIG 5] (Color online) Interface energy of Si wafers bonded using Al_2O_3 deposited at 200°C as the intermediate layer and annealed at 300°C for different duration. There was no significant change in the interface energy when annealing time was increased from 1 hr to 20 hr.

[FIG 6] (Color online) Interface energy of Si-Si wafers bonded using Al_2O_3 as intermediate layer annealed at 300°C for 1 hr, benchmarked against direct Si-Si bonding annealed for 1 hr (Maszara et al.¹) and 100 hr (Tong et al.²)

[FIG 7] (Color online) Images of 2-inch bonded wafers. (a) InP bonded to SiO_2 on Si and (b) GaAs bonded to Sapphire after a III-V substrate etch process.

[FIG 8] Cross-section SEM image of a bonded interface. Patterned SiO_2 on Si using dry etch was bonded to InP wafer with Al_2O_3 as the intermediate layer.